Multifractal spectra of chemical fields in fluid flows

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In the filamental phase of reactions embedded in fluid flows, where the concentration distribution is strongly fluctuating, we show that a chemical measure can be defined based on the absolute value of the concentration gradients. We express the generalized dimensions in terms of the roughness exponents of the structure functions as well as of the cancellation exponents of the chemical concentration. This measure is of basically different character than the natural distribution of the passive advection. It is similar to the SRB measures of dissipative systems, although the advection problem is area preserving. This approach is shown to be a useful tool in analyzing sea surface temperature anomalies.

I. INTRODUCTION

There has been an increasing interest in "active chaotic flows," i.e., in the problem of how the outcome of chemical or biological reactions can be influenced by the presence of hydrodynamical currents which generate chaotic particle advection [1]. The topic is of great practical importance ranging from chemical engineering and combustion [2] to geophysical problems like plankton blooms [3,4] or ozone depletion [5,6].

Earlier approaches have treated the problem of reactions in open and in closed flows separately. The reason is that in the open case there is a well defined fractal character (the unstable manifold of the chaotic set is not space filling) already in the passive advection problem due to the flux of fluid flowing through the system, and the product distribution is a fractal of the same dimension as the unstable manifold of the reaction free flow [7–10]. The fractality originates here from the advection dynamics, and the chemical products merely accumulate on it, as if it were a catalyst. In closed flows, however, the passive advection spreads over the fluid (the unstable filamentation is space filling) and so does the product, too, in the active case. For certain parameters the distribution is smooth, for others, however, it might become filamental (a smooth-filamental transion [12,13] can take place). In the filamental case the concentration distribution is rough. In the limit of weak diffusion, it is nondifferentiable along the stable foliation. This roughness is characterized by Hölder exponents or structure functions [11–15]. The concentration distribution is a self-affine function whose support is, however, not a fractal (rather the space of the flow).

Our aim is here to show that one can associate a fractal (more precisely multifractal) chemical distribution to the filamental phase. This is achieved by defining a chemical measure based on the absolute value of the concentration gradients. This measure is of basically different character than the natural distribution of the passive advection. It is similar to the SRB measures of dissipative systems [16] which is a multifractal distribution on chaotic attractors, although the advection problem is area preserving due to the incompressibility of the flow. Thus, the information dimen-

sion of the chemical reaction is in general less than that of the space of the flow, with an uniform natural distribution. This implies that *typical* concentration gradients can be found on a fractal subset of the full space only. More generally, we show here that in closed flows the chemical reaction itself produces the (multi)fractality and selects its dimension(s).

We first characterize the rough distribution in terms of cancellation exponents of sign-singular measure [17–20] which turn out to be equivalent with the use of structure functions. Next, based on the way of how a probability measure can be associated with a sign-singular one [20], we define the chemical measure and express its multifractal dimensions in terms of the cancellation and roughness exponents. Similar relations have already been found for the rough velocity fluctuations of fully developed turbulence [21], and the density fluctuations of a passive scalar [22]. The novel feature of our result is that it holds for a chemically *active* scalar.

The paper is organized as follows. In Sec. II we present the model and review previous results. In Sec. III the concentration distribution is characterized in terms of cancellation exponents and roughness exponents. We derive a relation between the generalized dimensions of the chemical measure and the above mentioned exponents. The validity of this relation is checked in numerical simulations. An application to geophysical data is discussed in Sec. IV. A brief summary is given in the concluding Sec. V.

II. THE MODEL

We consider the case of a single linearly decaying chemical substance advected by a time dependent two-dimensional closed flow. Although this seems to be a very special case, similar behavior appears for chemical reaction systems with a linearly stable equilibrium state [11,14]. A space dependent source $S(\mathbf{r})$ of the substance is also included, to maintain a nontrivial concentration field in a steady state.

The concentration field, $C(\mathbf{r})$, is governed by the partial differential equation:

$$\frac{\partial C(\mathbf{r})}{\partial t} + \mathbf{u}(\mathbf{r},t) \cdot \boldsymbol{\nabla} C(\mathbf{r}) = S(\mathbf{r}) - bC(\mathbf{r}) + D\Delta C(\mathbf{r}),$$

$$(2.1)$$

where $\mathbf{u}(\mathbf{r},t)$ is the known flow field, b the decay rate, and D the diffusion coefficient. For a general decaying reaction b is the absolute value of the Lyapunov exponent associated with the chemical dynamical subsystem.

For the flow we take a simple time periodic velocity field [23]. It is defined on a square of linear size $L=1$, with periodic boundary conditions, by

$$u_x(x,y,t) = 2U\Theta\left(\tfrac{1}{2} - t \bmod 1\right)\sin(2\pi y),$$

$$u_y(x,y,t) = 2U\Theta\left(t \bmod 1 - \tfrac{1}{2}\right)\sin(2\pi x), \qquad (2.2)$$

where $\Theta(x)$ is the Heaviside step function. In our simulations $U=1.2$, which produces a flow with a single connected chaotic region in the advection dynamics. The value of the numerically obtained Lyapunov exponent over one period, the time unit, is $\lambda \approx 2.7$. Trajectories on a rectangular grid were calculated and used to obtain the chemical field at each point for the linearly decaying reaction with the source term $S(x,y)=0.35[1+\sin(2\pi x)\sin(2\pi y)]$. We consider the situation when the diffusive transport is much weaker than the advective one: $D < LU$ (the Péclet number is large).

Depending on the relative strength of the chemical reaction and advection, two qualitatively different phases can appear in the steady state: the concentration field can be smooth or rough [11–15], as the result of the competition between two effects.

One effect is that of the chaotic flow which in the absence of reaction induces singular concentration patterns. A fluid domain becomes stretched in long and thin filaments, and is also repeatedly folded, producing a cascade of the inhomogeneities from the scale given by the source term toward smaller scales. The other effect is due to the stable chemical dynamics, which in a homogeneous environment tends to relax to a constant equilibrium distribution given by the fixed point of the reaction.

In the case of a slow chemical decay, filaments of high concentration persist for arbitrarily long times aligned with the unstable foliation of the flow (Fig. 1). If the decay of the chemical dynamics is faster than the dispersion of the chaotic trajectories in the physical space, irregularities of the field decrease and finally a smooth distribution is obtained. When the two effects become balanced, a smooth-filamental phase transition [12,13] takes place in the system, a macroscopic effect which affects the overall appearance of the chemical field.

In the filamental phase, the steady chemical field $C(\mathbf{r})$ is rough, it is a self-affine function. Thus, the Hölder exponent (α), given by the scaling of the concentration differences between two spatially close points:

$$|\delta C(\delta\mathbf{r})| \equiv |C(\mathbf{r}+\delta\mathbf{r}) - C(\mathbf{r})| \sim |\delta\mathbf{r}|^{\alpha}, \qquad (2.3)$$

serves as a proper tool for a quantitative characterization of the steady state concentration distributions.
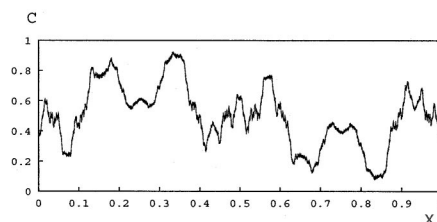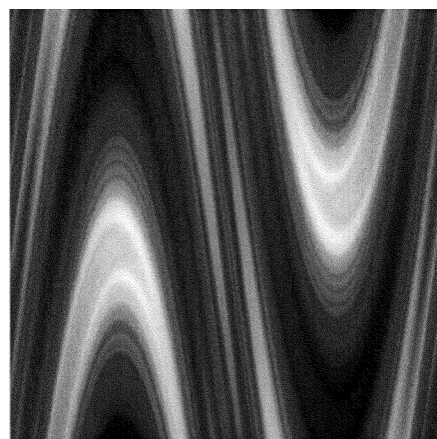

FIG. 1. Snapshot of the chemical pattern of the decaying substance under the chaotic flow (2.2) on the unit square in the (x,y) plane, for decay rate $b=2.4$, and with source $S=0.35[1+\sin(2\pi x)\sin(2\pi y)]$. Darker levels indicate smaller concentrations. Since $\lambda=2.7 > b$, a filamental structure is seen. The lower figure shows a cut of the concentration distribution taken at $y=0.25$.

In first approximation when the local Lyapunov exponents of the advection dynamics are assumed to be all the same (so-called monoaffine case) [12,13], one finds that

$$\alpha = \min\left\{\frac{b}{\lambda}, 1\right\}, \qquad (2.4)$$

where λ is the average positive Lyapunov exponent of the flow. This shows that the transition between the filamental (rough) and smooth concentration fields takes place when the Lyapunov exponents of the chemical and advection dynamics are equal in absolute value.

In order to take into account the fluctuations in the Lyapunov exponents, similarly, one can define a structure function characterizing the concentration distribution of the chemical field as

$$S_q(\delta\mathbf{r}) = \langle|\delta C(\delta\mathbf{r})|^q\rangle \sim |\delta\mathbf{r}|^{\zeta_q}, \qquad (2.5)$$

where the average denoted by $\langle\rangle$ is taken over different spatial regions. Above a certain cutoff scale (ε^*) the Hölder exponent (α) and the roughness exponents (ζ_q) are not expected to depend on the diffusion coefficient D.

In monoaffine approximation $\zeta_q = qb/\lambda = q\alpha$. In general, however, ζ_q is a nonlinear function of q. The multiscaling is determined by the distribution of the local Lyapunov exponents of the flow, since fractal sets with Lyapunov exponents different from λ dominate the scaling of the different q-order moments of δC.
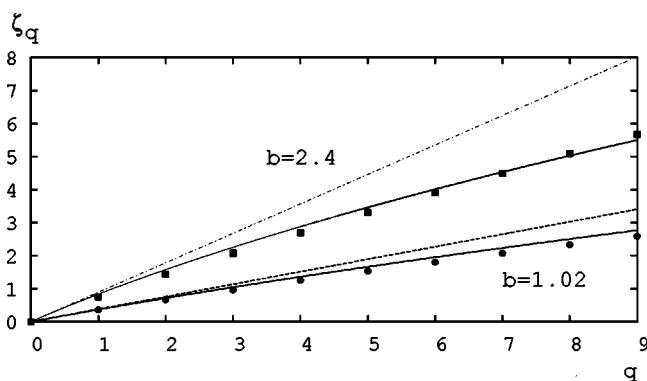
FIG. 2. The dependence of the scaling exponent ζ_q on q with reaction rates $b=1.02$ (circles) and $b=2.4$ (squares) for the reaction shown in Fig. 1. The continuous lines represent the theoretical prediction (2.8). $\Delta \approx 0.45$ is obtained by fitting Eq. (2.8) to the numerically obtained values of ζ_q. The monoaffine scaling $\zeta_q = q\alpha$ is represented by the dashed lines, where $\alpha = b/\lambda$ with $\lambda = 2.7$.

In the case when the probability distribution $P(\lambda')$ of the local Lyapunov exponents λ' is Gaussian, i.e., when

$$P(\lambda') \sim e^{-G(\lambda')t} \tag{2.6}$$

and

$$G(\lambda') = \frac{(\lambda' - \lambda)^2}{2\Delta}, \tag{2.7}$$

an explicit form can be derived [11,13]:

$$\zeta_q(b) = \sqrt{\left(\frac{\lambda}{\Delta}\right)^2 + \frac{2qb}{\Delta}} - \frac{\lambda}{\Delta}. \tag{2.8}$$

For small variances Δ of the Lyapunov distribution ($\Delta \ll \lambda^2/b$)

$$\zeta_q(b) = q\frac{b}{\lambda} - \frac{q^2}{2}\frac{b^2\Delta}{\lambda^3} + \frac{q^3}{2}\frac{b^3\Delta^2}{\lambda^5} + \cdots, \tag{2.9}$$

thus the deviation from a dynamically monofractal behavior produces a deviation from the monoaffine scaling.

The numerically obtained values of ζ_q are plotted in Fig. 2 along with the curve (2.8), and the monoaffine scaling exponents $\zeta_q = q\alpha$ with $\alpha = b/\lambda$, where $\alpha = 0.378$ and $\alpha = 0.889$ for the reaction rates $b=1.02$ and $b=2.4$, respectively. For comparison, the measured values of the first order roughness exponent are $\zeta_1 = 0.352$ and $\zeta_1 = 0.755$.

III. MULTIFRACTAL PROPERTIES OF THE CONCENTRATION DISTRIBUTION

In the following we define a chemical measure based on the concentration gradients. The major problem in finding this measure is, however, that the concentration gradients change sign, and cannot define a probability measure since the latter is positive by definition.

A. Sign singular measures: The cancellation exponent

Sign singular measures are distributions whose integrals change sign on arbitrary fine scales (above a cutoff scale ε^*)

[17–20]. A sign singular measure is a specific situation that cannot be taken for granted for all measures on data set. In order to decide if a measure is sign singular, one has to check if it possesses a positive cancellation exponent (see below). We show that the concentration increments in the filamental phase form a sign singular measure. The absolute value of this measure, however, can be used to define probability measures and we also show how the fractal properties of these measures are related to the cancellation exponents [18,20] characterizing the sign singular concentration distribution, and to the roughness exponents ζ_q of the structure function.

A measure is called sign singular, if it fulfills the property that the sum of the measure's absolute values in boxes of linear size ε diverges according to a power law for $\varepsilon \to 0$. The cancellation exponent $\kappa > 0$ of a sign singular measure μ defined on an interval is determined by computing the measures $\mu_i(\varepsilon)$ of ε-intervals via the relation

$$\sum_i |\mu_i(\varepsilon)| \sim \varepsilon^{-\kappa}, \tag{3.1}$$

for sufficiently fine resolutions above the crossover scale ε^*. The cancellation exponent thus measures how rapidly the derivative of the distribution changes sign with the resolution. Since the number of intervals is proportional to $1/\varepsilon$, this relation can also be written as

$$\langle |\mu(\varepsilon)| \rangle \sim \varepsilon^{1-\kappa}. \tag{3.2}$$

The average value (taken with a uniform distribution along an interval) of the modulus of the sign singular measure thus scales with the power $1-\kappa$ of the resolution.

The order q cancellation exponent κ_q is defined [20] as

$$\sum_i |\mu_i(\varepsilon)|^q \sim \varepsilon^{-\kappa_q}. \tag{3.3}$$

A nontrivial κ_q spectrum characterizes the inhomogeneity of the cancellation effect in the distribution, in an analogous way as the generalized dimensions characterize the inhomogeneous scaling in fractal probability measures. In terms of an average

$$\langle |\mu(\varepsilon)|^q \rangle \sim \varepsilon^{1-\kappa_q}. \tag{3.4}$$

In general, the chemical concentration field $C(x)$ along a line transversal to the local unstable foliation of the flow is typically an irregularly changing positive and bounded function of the location x. Its derivative $\partial C/\partial x$ changes sign on any scale down to a cutoff length ε^* set by diffusion. A sign singular chemical measure associated with an interval of length ε with x_i as its left corner, can thus be defined as

$$\mu_i(\varepsilon) = \int_{x_i}^{x_i+\varepsilon} \frac{\partial C}{\partial x} dx, \tag{3.5}$$

and it possesses a cancellation exponent spectrum κ_q. Since the absolute value of the sign singular measure is by definition $|\mu_i(\varepsilon)| = |C(x_i+\varepsilon) - C(x_i)|$, the structure function S_q given by Eq. (2.5) can be expressed as

$$S_q = \langle |\mu(\varepsilon)|^q \rangle, \qquad (3.6)$$

which scales as $S_q \sim \varepsilon^{\zeta_q}$. Therefore, in view of Eq. (3.4),

$$\zeta_q = 1 - \kappa_q. \qquad (3.7)$$

We have thus obtained an explicit relation between roughness exponents and cancellation exponents (see also [22]). This implies that $\kappa > 0$ whenever $\zeta_1 < 1$, i.e., the concentration increments form then a sign singular measure. On the other hand, $\zeta_1 < 1$ is the condition for a filamental phase, which is an extension of $\alpha < 1$ in Eq. (2.4) for flows with a Lyapunov exponent distribution.

B. Generalized dimensions and roughness exponents

Consider now, in the spirit of [20], the positive measure

$$\bar{\mu}_i(\varepsilon) = \frac{\int_{x_i}^{x_i + \varepsilon} \left| \frac{\partial C}{\partial x} \right| dx}{\int \left| \frac{\partial C}{\partial x} \right| dx} \qquad (3.8)$$

based on the absolute value of concentration gradients. Due to the denominator, this is normalized, and can be considered as a probability measure. Its generalized dimensions D_q follow from the relation [24]

$$\sum_i \bar{\mu}_i(\varepsilon)^q \sim \varepsilon^{(D_q - 1)(q-1)}. \qquad (3.9)$$

It has been shown in [20] that these dimensions can be expressed by means of the cancellation exponent of the sign singular measure (3.5). To see this, let us imagine that the interval of length ε appearing in Eq. (3.8) is decomposed into several intervals whose length is the lower cutoff ε^*. On this scale, the concentration gradient does not change sign any longer, therefore the measure $\bar{\mu}$ of these intervals is the same as the absolute value of their sign singular measure μ. Thus

$$\bar{\mu}_i(\varepsilon) = \frac{\sum_i' |\mu(\varepsilon^*)|}{\sum_i |\mu(\varepsilon^*)|}, \qquad (3.10)$$

where Σ' denotes that the summation is only over the small intervals which fall into the large one of length ε. In order to express the right-hand side via the sign singular measure of this whole interval, we observe first that the definition (3.1) of the cancellation exponent can equivalently be written as

$$\sum_j |\mu_j(\varepsilon)| \varepsilon^\kappa = \sum_i |\mu_i(\varepsilon')| \varepsilon'^\kappa \qquad (3.11)$$

for any resolution ε and ε' covering the same interval of some length ε_0. Applying this relation to $\varepsilon' = \varepsilon^*$, and to $\varepsilon_0 = \varepsilon$, when in the latter case there is only one term on the right-hand side, we find

$$\sum_i' |\mu_i(\varepsilon^*)| = |\mu_i(\varepsilon)| \left(\frac{\varepsilon}{\varepsilon^*} \right)^\kappa. \qquad (3.12)$$

Since the denominator of Eq. (3.10) scales as $(\varepsilon^*)^{-\kappa}$,

$$\bar{\mu}_i(\varepsilon) \sim |\mu(\varepsilon)| \varepsilon^\kappa, \qquad (3.13)$$

i.e., the probability measure of an interval is the absolute value of the sign singular measure of the same interval multiplied by power κ of the resolution. This relation is valid only with the condition $\varepsilon \gg \varepsilon^*$, since the validity of Eq. (3.12) is restricted to cases when the number of terms in the sum on the left-hand side is large. Substituting Eq. (3.13) into the definition (3.9) of the generalized dimensions, and using Eq. (3.3) we obtain

$$D_q = 1 + \frac{\kappa q - \kappa_q}{q - 1}. \qquad (3.14)$$

In terms of the scaling exponents [see Eq. (3.7)]

$$D_q(b) = 1 + \frac{q[1 - \zeta_1(b)] - 1 + \zeta_q(b)}{q - 1}. \qquad (3.15)$$

A similar relation has been derived between the structure function exponent and the generalized dimension of the velocity fluctuations in fully developed 3D turbulence [21], and of a diffusively decaying passive tracer in quasigeostrophic turbulence [22]. In our case the presence of an asymptotic steady state is due to the chemical decay and the source, and the roughness is generated by a smooth flow.

The generalized dimensions are functions of the chemical reaction rate b. Note that the parameters of the source distribution do not appear, only its smoothness is important. D_q is also independent of the diffusion coefficient, since the roughness exponents are. Although the example considered is a linearly decaying reaction, the phenomenology is the same for all chemical dynamics converging towards a stable state, as justifed in [11,14]. Therefore Eq. (3.15) is expected to be valid for general stable chemical dynamics when b is the decay rate (the chemical Lyapunov exponent) characterizing the convergence towards the chemical attractor.

As a conclusion, we can claim that the self-affine distribution of the concentration gradients characterized by the scaling exponents determines the multifractal properties of the positive measure based on the absolute value of the concentration gradients. In other words, the result shows that the chemical reaction puts on the passive advection problem a measure different from its natural measure (which is characterized in closed flows by $D_q^{(\text{passive})} \equiv 2$). In the smooth phase when $\zeta_q = q$, we find $D_q \equiv 2$. The presence of a nontrivial chemical multifractal spectrum thus implies the roughness of the concentration distribution.

Based on relation (2.9) we obtain for the multifractal spectrum

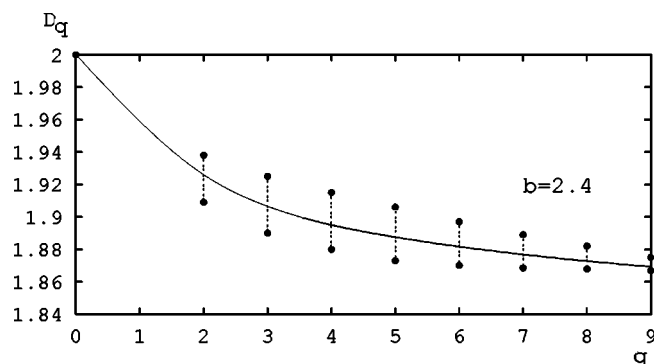$$D_q(b) = 2 - \frac{q}{2} \frac{b^2 \Delta}{\lambda^3} + \frac{1}{2} q(q+1) \frac{b^3 \Delta^2}{\lambda^5} + \cdots. \qquad (3.16)$$

FIG. 3. The multifractal spectrum of the chemical measure for $b=2.4$. The error bars are obtained as the differences of the slopes in the ε range $[1/1024, 1/256]$ and $[1/256, 1/64]$. The continuous line represents expression (3.15) based on the measured exponents ζ_q presented in Fig. 2.

An important special case of generalized dimensions is the information dimension D_1, since its deviation from the fractal dimension already implies multifractality (and Kaplan-Yorke type relations only contain the information dimension). We obtain from Eq. (3.14)

$$
\begin{aligned}
D_1(b) &= 1 + \kappa_1 - \left.\frac{d\kappa_q}{dq}\right|_{q=1} \\
&= 2 - \left(\zeta_1 - \left.\frac{d\zeta_q}{dq}\right|_{q=1}\right) \\
&\approx 2 - \frac{1}{2}\frac{b^2\Delta}{\lambda^3} + \frac{b^3\Delta^2}{\lambda^5} + \cdots.
\end{aligned} \qquad (3.17)
$$

Since the slope $d\zeta_1/dq$ at $q=1$ is less than ζ_1 itself (cf. Fig. 2), the information dimension $D_1(b)$ is indeed less than $D_0(b)=2$. Thus, the measure based on the modulus of the concentration increments is a multifractal measure.

C. Numerical results

Since the validity of Eq. (3.15) is constrained on the one hand to $\varepsilon \gg \varepsilon^*$ to assure the statistical scaling behavior of the sign singular measure with power κ, and on the other hand to $\varepsilon \ll 1$ (1 is the system size) to produce the scaling with power D_q, the scaling behavior is restricted to a short range in ε only. We determine the measure at the fixed value $\varepsilon^* = 1/4000$, and evaluate $\bar{\mu}(\varepsilon)$ in the range $1/4000 < \varepsilon < 1/64$.

The range $\varepsilon \in [1/1024, 1/64]$ seems to be suitable for dimension measurements, in agreement with the theoretical arguments which require $\varepsilon^* \ll \varepsilon \ll 1$. The average slopes of the graphs $\ln \Sigma \bar{\mu}_i^q$ vs $\ln \varepsilon$ in this range give the values D_q, which are plotted in Fig. 3 along with the curve (3.15) [25].

An evaluation of the numerically obtained scaling exponents results in the derivative $d\zeta_q/dq|_{q=1}=0.71$ for the case $b=2.4$. Together with the first order roughness exponent ζ_1

$=0.755$, Eq. (3.17) gives $D_1=1.955$. The same information dimension value can be read off from the graph of D_q.

IV. APPLICATION TO SEA SURFACE TEMPERATURE DATA

An appealing application of the above ideas is the investigation of the sea surface temperature (SST) anomalies. SST can be considered in first approximation as a linearly decaying substance towards its local average value. Abraham and Bowen [15] analyzed satellite images with 1 km resolution of the SST distribution in a rectangular region of the size of few hundred kilometers in the Tasman sea, and found filamental distribution. Using estimated sea surface velocity data obtained from correlations in sequential SST images, they have shown that the advection dynamics is characterized by a spectrum of Lyapunov exponents whose distribution is of the type of Eqs. (2.6) and (2.7), with an average Lyapunov exponent and variance $\lambda=4\times10^{-7}$ 1/s and $\Delta=4.5\times10^{-7}$ 1/s, respectively. The decay rate of the SST anomalies was found to be $b=0.03/$day. The authors were able to determine the roughness exponents ζ_q of the measured SST distribution in this time dependent flow. The spectrum ζ_q was shown to obey the form (2.8) with good accuracy. Extracting the values $\zeta_1=0.65$ and $d\zeta_q/dq|_{q=1}=0.483$ from the data of [15], we obtain from Eq. (3.17) the information dimension of the measure based on the SST gradients as $D_1=1.833$. It is remarkable that a clean fractal dimension less than 2 can be associated with typical SST gradients. Since the data fulfill both $\kappa \equiv \kappa_1=1-\zeta_1>0$ and $D_1<2=D_0$, we conclude that the SST increments form a sign singular measure, and their modulus (after normalization) yields a multifractal distribution.

Furthermore, this oceanic flow provides an example of two-dimensional turbulence, in which the time dependence of the locally smooth velocity field does not have any kind of periodicity. This illustrates that the multifractal properties of decaying fields can also be obtained in realistic situations, when the flow is chaotically time dependent.

We note, that in general the oceanic velocity field cannot be obtained directly by remote sensing. Therefore, it is difficult to quantify oceanic stirring by Lyapunov exponents and their distribution. However, the distribution of SST is accessible with relatively high resolution. Thus, when SST increments form a sign singular measure, the relationship between cancellation exponents and Lyapunov exponents could be used to obtain information about the advection dynamics even without access to the velocity field.

V. CONCLUSIONS

Reactive flows can be characterized by various multiscaling properties, described by apparently different exponents (κ_q, ζ_q, D_q) evaluated in this paper, but, in fact, they represent equivalent descriptions, since all of them are derived from the rough concentration field $C(\mathbf{r})$. These exponents

could be used to characterize statistical scaling properties of the chemical concentration fluctuations in measured data obtained from geophysical observations or laboratory experiments. They provide a way to extract information beyond the mean concentrations of the different chemical components, that could be used, for example, as a tool for monitoring changes in the complex chemical or transport processes involved.

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[25] Examining the scaling of another positive entity: $\bar{\mu}'_i(\varepsilon) = |\int_{x_i}^{x_i+\varepsilon}(\partial C/\partial x)dx|/\int|\partial C/\partial x|dx$, and its dimension defined by $\Sigma_i\bar{\mu}'_i(\varepsilon)^q \sim \varepsilon^{(D'_q-1)(q-1)}$, we find for D'_q the same relation as Eq. (3.15) directly from Eq. (3.5). In this case the suitable ε range is no longer restricted by Eq. (3.11) to a limited region in ε and the measurements' errors become smaller.